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SOIL RADIONUCLIDE ADSORPTION AND PARTICULATE FILTRATION IN AN N-AREA SOIL

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by

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Water and Wastewater Research

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SOIL RADIONUCLIDE ADSORPTION AND PARTICULATE FILTRATION IN AN N-AREA SOIL

B. L. Carlile and B. F. Hajek

INTRODUCTION

This report presents interim experimental results of an investigation requested by the N-Reactor Department, Richland, Washington. The purpose of this research was to evaluate the extent of radionuclide movements, both ionic and particulate, from a ground disposal facility designed for disposal of radioactive waste coolant generated in the event of a reactor primary loop failure incident. This report summarizes Phase I of the study - the characterization of cesium and strontium mobility at less than 10^{-2} percent breakthrough.

In studies conducted previously for the N-Reactor Department (2), it was shown, by some extrapolation of data, that 12 column volumes of radioactive waste coolant could be disposed to a ground disposal facility with a relative trace strontium breakthrough of 10^{-2} percent (C/Co x 100). However, subsequent estimates of influent radionuclide concentrations made by N-Reactor personnel showed that an activity reduction to 10^{-4} percent of influent concentration was needed. This necessitated further extrapolation of experimental data which showed an ultimate ground disposal capacity of 6 column volumes.

Where field predictions of radionuclide breakthrough into groundwater are made from soil column data, some extrapolation of the analytical data is possible since a log-probability plot of C/Co versus total column volumes is linear to some lower limiting value of C/Co (2). However, since this theoretical approach is based on a mass action formulation of ion exchange, many uncertainties are associated with extrapolating very far beyond experimental data. Therefore, an exploratory study was conducted in which IRP loop wastewater was permeated through a soil column. This study indicated a strontium-90 relative concentration breakthrough of 10^{-2} percent occurring at 4 column volumes throughput.

However, the analytical limitations at this low concentration could account for some of the discrepancy in the data (5).

At this point it was evident that additional studies were necessary to confirm the adequacy of ground disposal for this type of wastewater. The primary requirement for this study was a radionuclide activity high enough to obtain effluent concentrations which can be analyzed with confidence at a 10^{-2} to 10^{-4} percent activity reduction to determine if breakthrough data can reasonably be represented by a straight line log-probability curve at these lower limits.

SUMMARY

Laboratory soil column investigations with high activity cesium and strontium solutions show breakthrough values to be appreciably higher than previous extrapolated predictions for N-Area soils.

The experimental results showed an initial breakthrough of 10^{-1} percent occurring with both ^{137}Cs and ^{85}Sr at less than 0.1 column volume throughput. After this initial pulse through the column, activity leakage was sporadic, ranging from background to values as great as 1.0 percent for each sample of effluent. This random leakage continued until approximately 5 column volumes of solution had permeated through the column. At this point the activity leakage was reduced to essentially the lower limits of analytical detection (10^{-3}%).

From the breakthrough pattern obtained in the initial phase of this study, it was evident that some phenomena other than ion exchange was responsible for this effluent activity. The three possible mechanisms of activity transportation are: ionic solution transport, diffusive movement, and colloidal or particulate migration. Since only the latter mechanism could result in the breakthrough pattern obtained in this study, the possibility of particulate migration was investigated.

Subsequent experiments showed that much of the activity in the eluting solution was associated with very fine colloids migrating through the column. Additional studies indicated most of these colloids were inorganic rather than organic in nature. Whether these particulates are very fine clays coming from the soil material itself or originating in the river water is speculatory at the present, although first indications point to the soil material as the source.

Preleaching the column with river water before addition of radioactivity reduced the activity leakage by a factor of 10 to 20. Some reduction in initial breakthrough was also accomplished by the addition of NaCl to the influent solution to aid in the flocculation of the fine colloids.

METHODS AND MATERIALS

SOIL COLUMNS

The cesium and strontium breakthrough resulting from the infiltration of a simulated waste liquid, was determined by the use of laboratory soil columns, through which the waste liquid was percolated and the effluent collected for analysis. The columns used in this study were 2 cm in diameter and 40 cm in length. Previous studies at Hanford have shown column diameter to be inconsequential and short column length to be conservative in estimating trace ionic breakthroughs in ground disposal systems (4).

The soil used in this study was obtained from test wells located in the vicinity proposed as a probable location for the disposal facility. A composite was made from soils obtained at various depths in these wells and a portion of this composite packed into the columns at approximately field bulk density (1.9gm/cc). Mechanical analysis of the soil material from different well sites and various depths indicated very little variation between samples making up the composite (1).

The waste liquid in this study was simulated by spiking raw river water with traces of strontium-85 or cesium-137. The activity in each was approximately 100,000 cpm/ml. Activity leakage through the column was determined by analysis of the effluent collected with an automatic fraction collector. Each one to two milliliters of effluent was collected and analyzed separately to determine a pattern for activity breakthrough.

For a confidence level of .95, significant detection at a 10^{-3} percent breakthrough factor was accomplished by taking long multiple counts of each sample coupled with equivalent background counts between samples.

PARTICULATE ANALYSIS

Effluent samples containing the greater amounts of activity were studied in further detail to verify presence of particulates. Each of these samples was centrifuged at high speeds and a small aliquot drawn from the upper portion of the solution and recounted. The fraction of activity centrifuged as particulates was determined from the fraction remaining in the liquid phase.

Additional information about the origin of the particulates carrying the activity was obtained by treating the samples with 30 percent H_2O_2 at pH7 and heating to approximately 70°C. The samples were again centrifuged and an aliquot drawn and counted as before. This treatment, at neutral pH, removes the organic matter without significantly altering soil mineral colloids (3). Any activity associated with organic colloids would be released to solution and counted in the solution phase.

REDUCING ACTIVITY TRANSPORT

After verification of particulate activity carriers was made, additional studies were conducted whereby soil columns were preleached with 1 column volume of river water before percolation with the waste solution. From porosity calculations for each column, the volume of river water retained in the columns after preleaching were determined. Since the percolating waste solution would displace this volume of solution ahead of it, corrections for column volume waste discharge could be accurately made.

Two additional columns were set up and leached with a simulated waste solution made 1M with sodium chloride. The purpose of this experiment was to study soil flocculation as a means of reducing activity leakage.

RESULTS AND DISCUSSION

The pattern of breakthrough for strontium and cesium through columns of N-Area soil is shown in Figures 1 and 2. The character of this breakthrough, plotted on logarithmic probability coordinates, is such that no extrapolation of data is possible, indicating some mode of action other than mass action ion exchange is responsible for this transport of activity through the columns.

Figures 3 and 4 show the same data plotted as accumulative breakthrough; i.e., C/Co represent the fraction of total accumulative effluent activity over total accumulative influent activity at each stage of leaching. These curves are a more meaningful representation of the data since this shows the relative concentration of radioactive strontium and cesium which would reach groundwater after disposal of equivalent field column volumes.

From the data it is evident that the disposal of any volume of waste solution of cesium and strontium sufficient to reach groundwater will exceed the required reduction in activity of 10^{-4} percent. Therefore, some pretreatment of the soil or waste solution is needed to effectively reduce this leakage in a ground disposal system.

Figure 5 shows the results of a centrifugation study made on the effluent samples highest in activity. This data confirms the postulation of activity being associated with very fine particulates migrating through the column.

These results indicate that 50 - 75 percent of the activity in the effluent can be removed by centrifugation and this value could be even greater since the centrifuge system used in this study only removed colloids greater than 0.05 microns in diameter. Fine clay colloids may range down to 0.001 microns in size and these would remain in suspension to be counted with the solution under the conditions used in this study.

Flushing the column with 1 column volume of river water before leaching the waste solution had the effect of reducing activity breakthrough by a factor of 10 to 20. Figures 6 and 7 show the results of this study. Preleaching the column with river water flushes a portion of those fine particulates from the column, reducing the number of colloids available for activity carriers. Continued preleaching with a total of 4 to 5 column volumes of river water should reduce breakthrough even more since colloid elution from the column essentially stopped at this point. However, preleaching a field disposal system with 4 to 5 column volumes of water would be an expensive and troublesome operation and impractical unless alternative treatments were not sufficient in reducing leakage.

Leaching a soil system with river water, low in ionic strength, has the effect of dispersing the soil into its individual fractions. This situation is created by a high degree of hydration and a high electrostatic charge on the soil particle due to the removal of cationic species from the exchange sites. Some treatment to effectively reduce this dispersion (and promote flocculation) should result in a significant decrease in migration of colloids through the column with an accompanying decrease in activity leakage.

A standard procedure for the flocculation of soil mineral colloids is the addition of a sodium salt in sufficient strength to result in an excess of free sodium ions. This can be shown by the following equilibrium equation:

$$NaX \rightleftarrows Na^+ + X^-$$

where X represents the soil colloid. When the sodium concentration is high, the equilibrium is shifted to the left, decreasing the negative charge on the particles, promoting flocculation.

Although this procedure is successful in promoting flocculation, it is undesirable from the standpoint of cesium and strontium adsorption. The excess sodium will compete with the cesium and strontium for adsorption sites on the soil colloid, thereby increasing the average migration rate of the cesium and strontium ions through the column. The net result would be a decrease in initial activity migration of colloids but a faster breakthrough due to ionic movements in solution.

There are several chemical additives available today which might be used to reduce dispersion without the undesirable effects of ion competition from sodium. Examples of such compounds are the polyelectrolytes and flocculating agents used in water filtration plants. The benefits of these materials in soil systems have not been proven at the present time, however.

CONCLUSIONS

The significant results of this study are summarized and listed as follows:

1. Breakthrough of cesium and strontium exceeding 10^{-4} percent occurs with the first fraction of solution throughput.
2. This breakthrough is not continuous but sporadic, varying from background levels to as high as 1.0 percent.
3. A substantial portion of this activity leakage is associated with fine inorganic colloids migrating through the column.
4. Preflushing the column with 1 column volume of river water before addition of the waste solution reduced breakthrough by a factor of 10 to 20.

5. The addition of sodium chloride to the waste solution reduced initial breakthrough by a factor of 10 but will have detrimental effects on ion adsorption.

6. Additional investigations will be necessary to determine the specific origin of the activity-carrying colloids and develop methods to reduce this initial breakthrough to acceptable levels.

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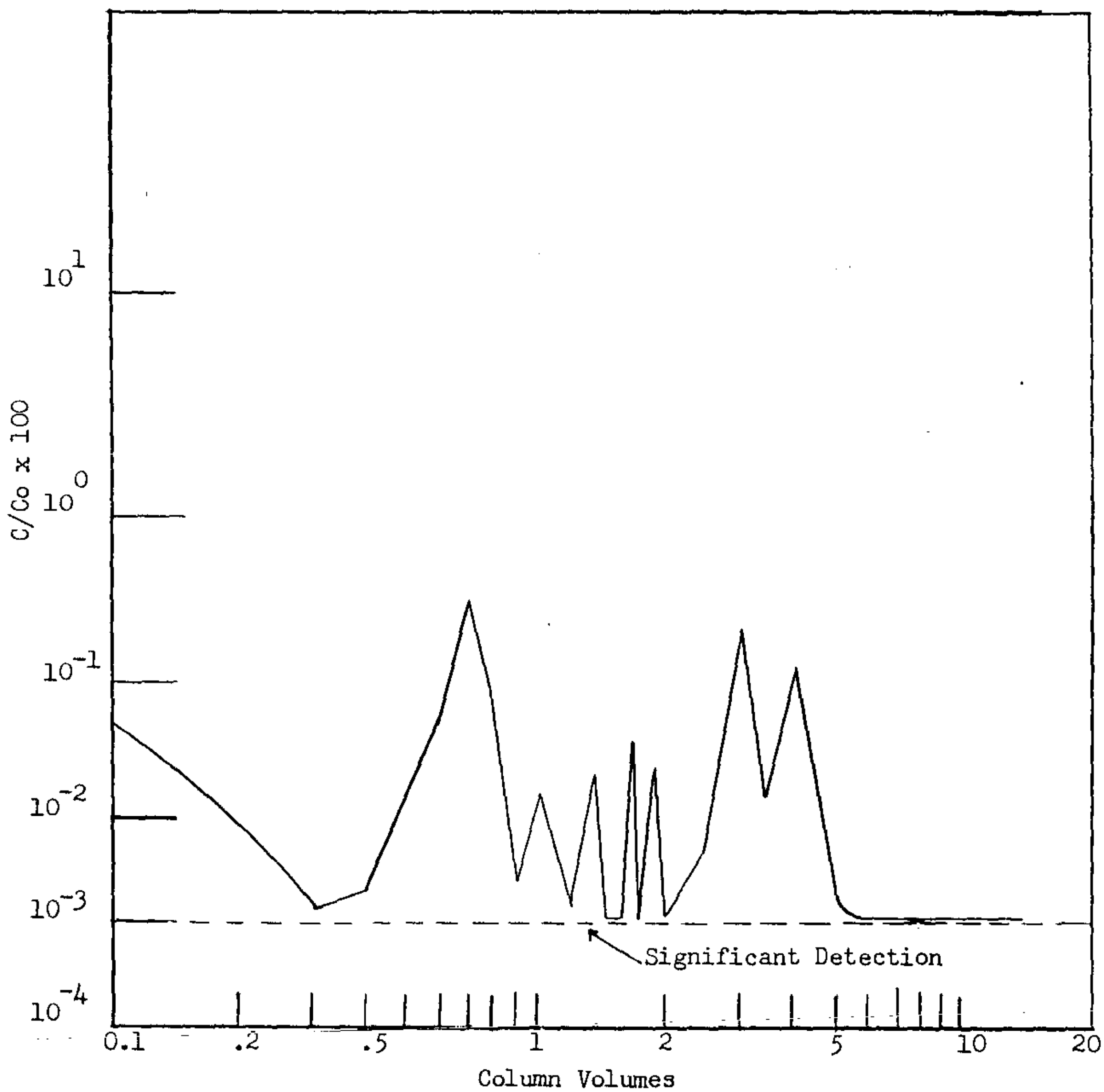


<u>FIGURE 1</u>

Cesium Breakthrough Curve (Error Function C/Co x 100 versus Log Column Volumes).

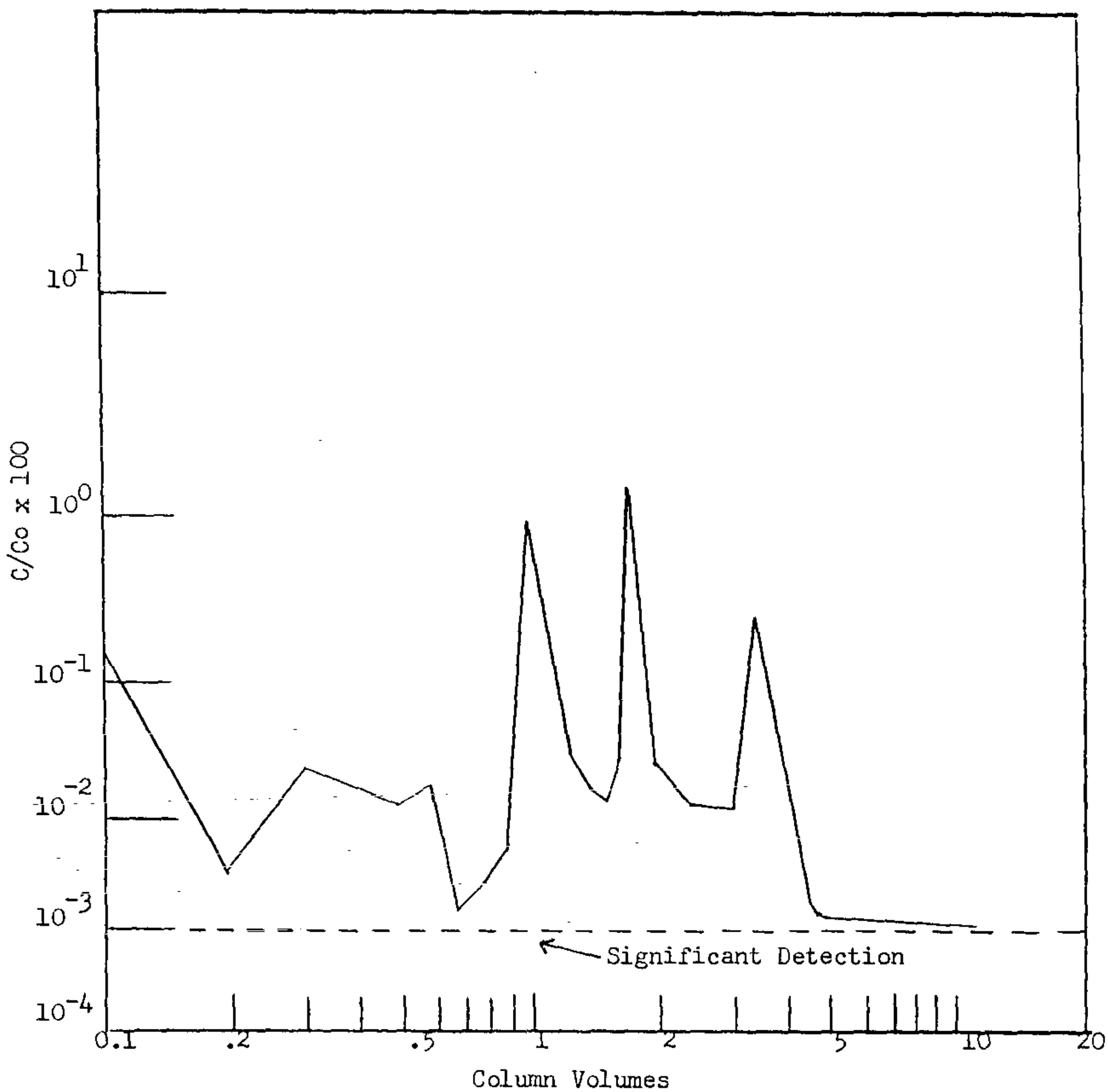
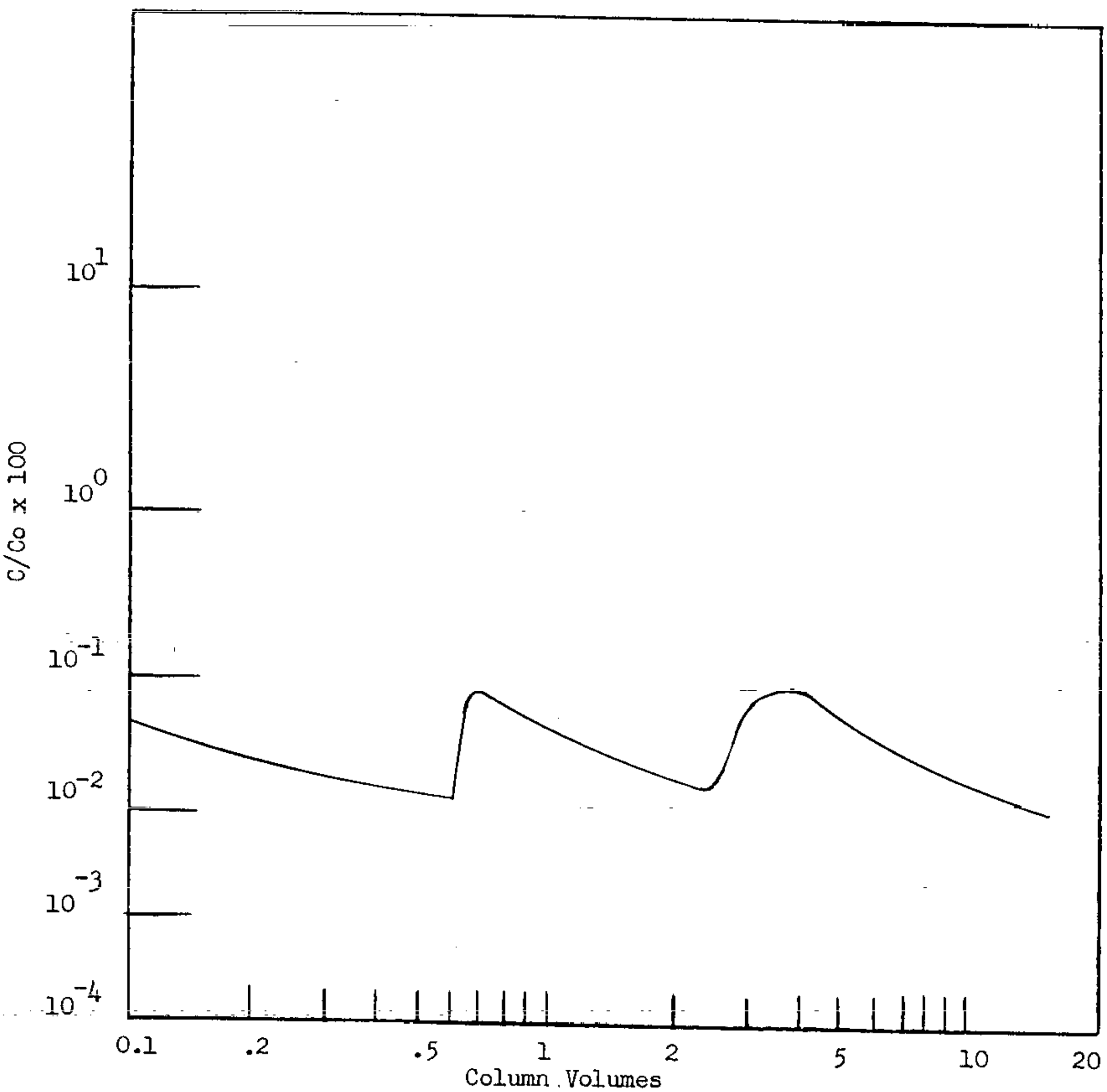


FIGURE 2

Strontium Breakthrough Curve (Error Function C/Co x 100 versus Log Column Volumes).



<u>FIGURE 3</u>

Accumulative Cesium Breakthrough Curve (Error Function C/Co x 100 versus Log Column Volumes).

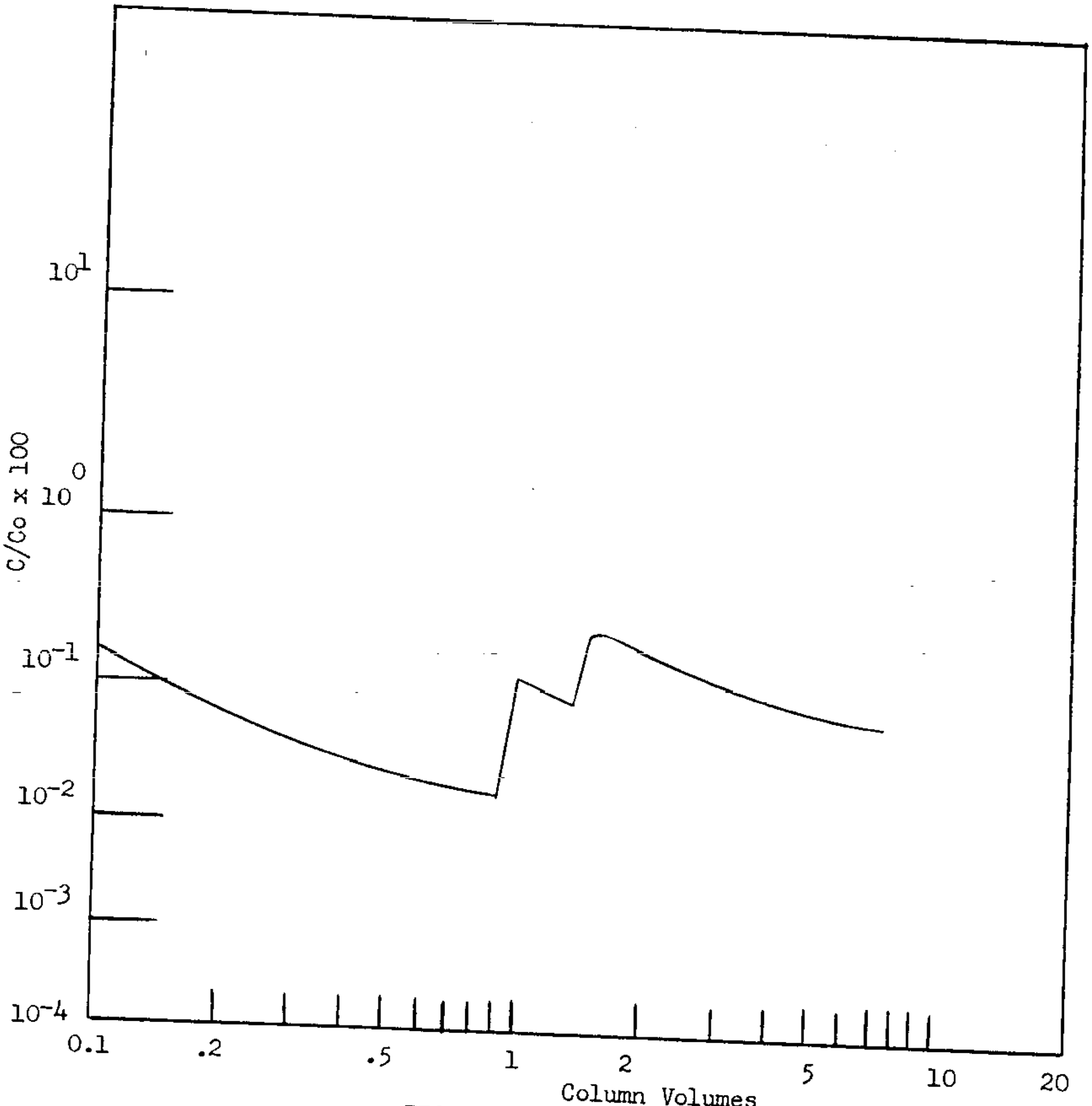


FIGURE 4

Accumulative Strontium Breakthrough Curve (Error Function C/Co x 100 versus Log Column Volumes).

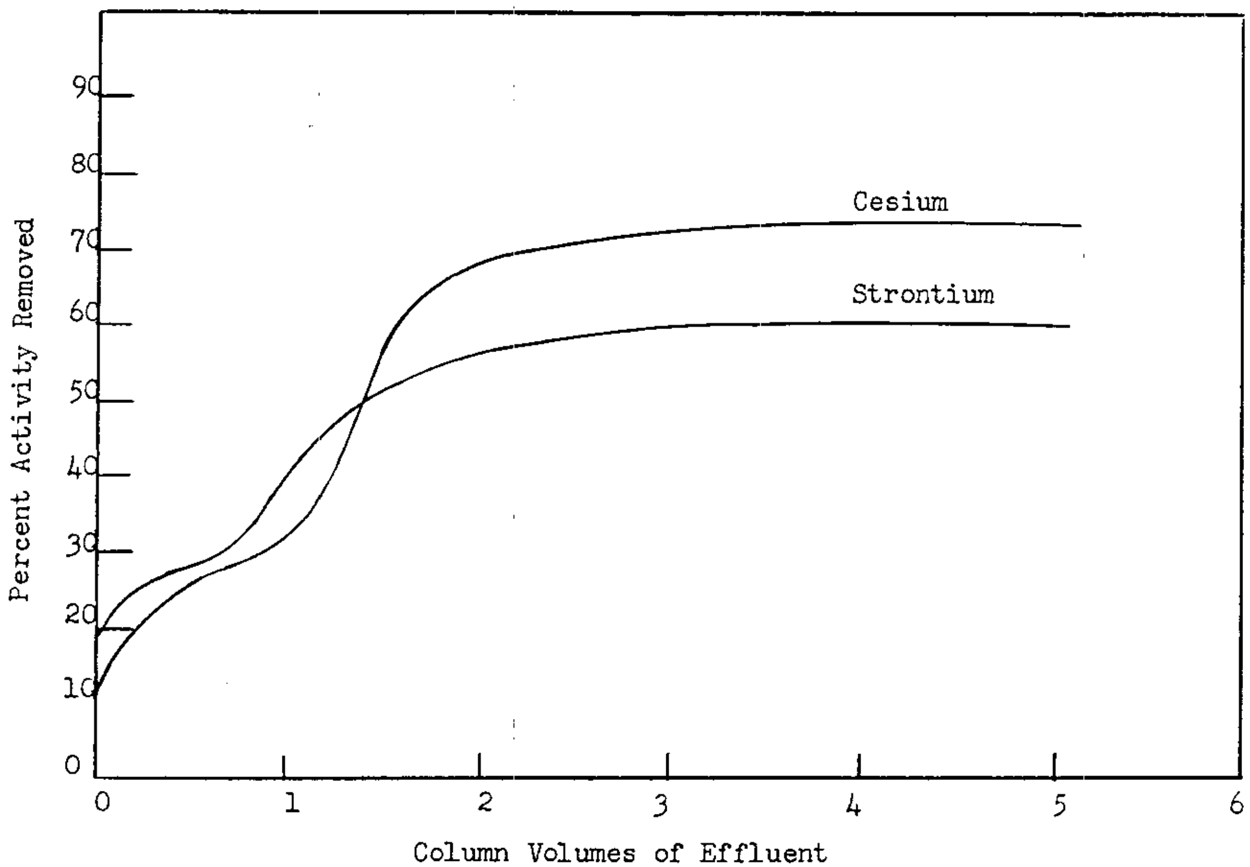


FIGURE 5

Reduction in Activity by Centrifugation of Effluent Solution

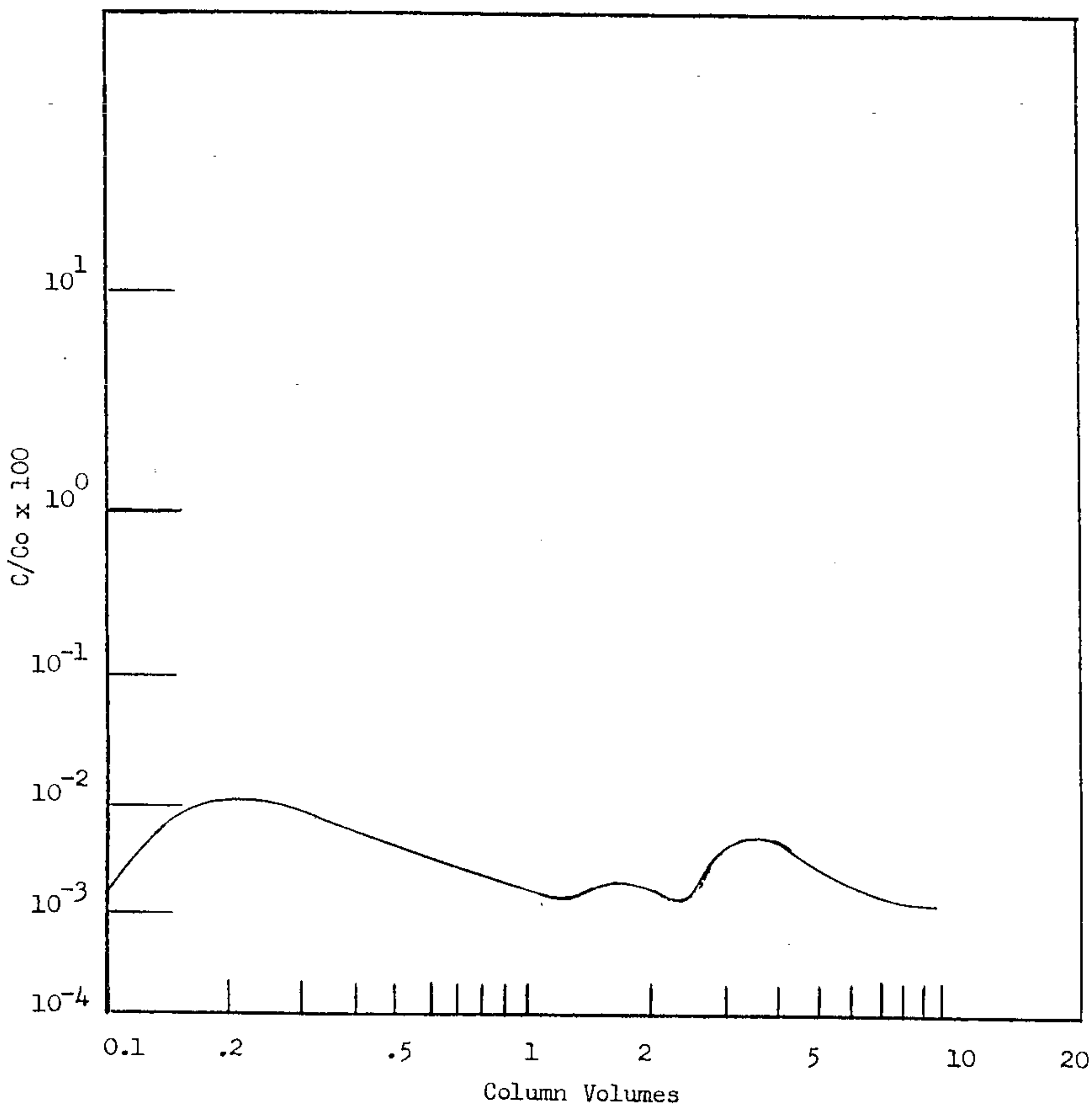


FIGURE 6

Accumulative Cesium Breakthrough Curve from Column Preflushed with River Water (Error Function C/Co x 100 versus Log Column Volumes)

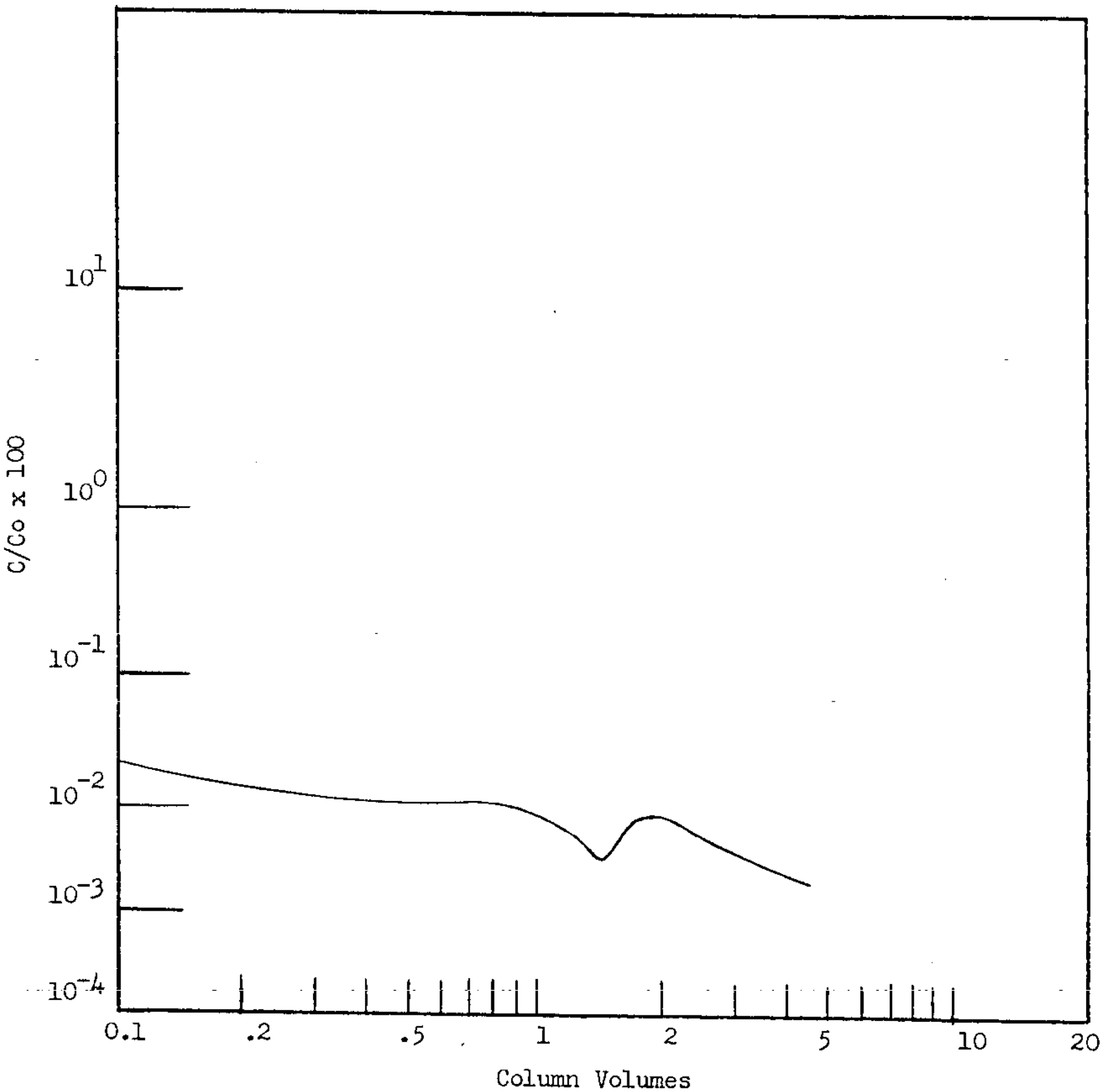


FIGURE 7

Accumulative Strontium Breakthrough Curve from Column Preflushed with River Water (Error Function C/Co x 100 versus Log Column Volumes)